ANL/CSE-13/8

Qualification Status of 6-gU/cm^3 U_3Si_2 Dispersion Targets for ^{99}Mo Production

Chemical Science and Engineering Division

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QUALIFICATION STATUS OF 6-gU/cm^3 U_3Si_2 DISPERSION TARGETS FOR ^{99}Mo PRODUCTION

1 INTRODUCTION

In October 1988 the U.S. Nuclear Regulatory Commission (NRC) approved the use of U_3Si_2-Al dispersion fuel in its licensed plate-type reactors at densities up to 4.8 gU/cm^3 and up to power densities and ^{235}U burnups typical of fuels tested in the Oak Ridge Research Reactor (ORR) (NRC 1988). Since that time, regulatory authorities in many other countries have approved use of U_3Si_2 plate-type fuel. The French research reactor fuel fabricator (CERCA) continued fabrication development and announced in 1992 its ability to provide U_3Si_2 dispersion fuels up to a density of 6.0 gU/cm^3 (Durand et al. 1993). Five fuel plates, each having uranium densities of 5.8 and 6.0 gU/cm^3, were irradiated in Siloe to a burnup of 55% (at.% ^{235}U) from 1995 to 1997, and two 5.8-gU/cm^3 fuel assemblies were irradiated in Osiris to a burnup of 74% in 1997 and 1998, all with very good results (Durand et al. 1998). These manufacturing developments and irradiations demonstrated that the fuel can be manufactured reliably and that it performs extremely well under irradiation; hence, by the definition of qualification given in *Guidelines for Qualification and Licensing of Very-High-Density Fuels* (IAEA in press), 6.0-gU/cm^3 U_3Si_2 dispersion fuel can be considered to be qualified for use under conditions that do not exceed those of the test irradiations.

A number of ^{99}Mo producers use targets that are small plates of dispersion fuel; for example, the Institute for Radioelements (IRE) and Covidien use highly enriched UAl_x and the ANSTO (Australian Nuclear Science and Technology Organization), CNEA (Argentina's Comisión Nacional de Energía Atómica), and NESCA (Nuclear Energy Corporation of South Africa) use low-enriched UAl_2. When changing target type, one must pay especial attention to processing aspects. Many years ago, Dr. Ali Sameh and colleagues developed a process at Karlsruhe appropriate for use with U_3Si_2 dispersion fuel plates (Sameh and Bertram-Berg 1993).

2 U_3Si_2 BEHAVIOR UNDER IRRADIATION

Kim et al. (2009) provides a comprehensive review of the irradiation behavior of U_3Si_2 dispersions under conditions applicable to most research reactors. Experiments have shown that U_3Si_2 dispersions are very stable under irradiation to high burnup as long as the interaction-product formed around the U_3Si_2 particles is in contact with the aluminum matrix. At any stage of irradiation, the amount of matrix remaining depends on the amount of interaction product produced, which, in turn, depends on the fission rate, the fuel temperature, and the length of time under irradiation. Figure 1 shows the effect of variations in the fission rate and the irradiation temperature on the fission gas morphology in U_3Si_2 fuel particles. It should be noted that the samples shown and discussed here came from a 75%-enriched, 4.7-gU/cm^3 fuel plate used to simulate high fission rates and fission densities typical of low-enriched U_3Si_2 fuel plates in high-flux reactors. It should also be noted that the fission gas bubbles appear only in the unreacted U_3Si_2 fuel and not in the interaction product. The fission densities of samples (a) and (b) are equivalent to ^{235}U burnups of ~62% and ~96% in LEU fuel; the fission density of sample (c) is higher than the ~5.8×10^{21} f/cm^3 that represents 100% burnup in low-enriched U_3Si_2 fuel. Fission gas bubbles are much larger in the higher-fission rate/temperature portions of the fuel plate; such large bubbles form by coalescence of smaller bubbles. The larger the bubble, the lower its surface tension, resulting in an increasing growth rate; if bubbles begin to interconnect from fuel particle to fuel particle, as is especially likely in high-density fuels, breakaway swelling, or pillowing, of a plate-type fuel has begun. Data from the many irradiation experiments analyzed in Kim et al. (2009) show that there is a threshold in a plot of fission density versus life-average fuel temperature below which fission gas bubbles remain below 5 μm in diameter, which is conservatively small enough to prevent bubble interconnection, and that there is a higher threshold that represents the conditions for large-bubble interconnection, as shown in Figure 2.

The failure mode discussed above is caused by the growth of large fission gas bubbles in the fuel particles and their subsequent interlinking. A different failure mode that can also result in fuel plate pillowing was identified for U_3Si_2 dispersion fuel from the results of very-high-temperature irradiations of highly enriched 4.6-gU/cm^3 U_3Si_2 dispersion samples carried out in the HFIR (High Flux Isotope Reactor) at Oak Ridge National Laboratory in support of the Advanced Neutron Source project (Copeland et al. 1995). The initial fission rates during this irradiation were between four and seven times the initial fission rates in the samples shown in Figure 1. The samples were small punchings from miniplates fabricated by the normal roll-bonding process, so they are typical of regular dispersion fuel plates. These samples were irradiated at a nearly constant temperature in spite of their high burnup because only 10% of the heat was produced by fission. In contrast to the highly irradiated samples shown in Figure 1, which exhibited large fission gas bubbles, the porosity in these samples, two of which are shown in Figure 3, appears to have been produced by fracturing of the interaction product. The spaces between the fracture surfaces are obviously filled with fission gas because the meat in the 425°C samples has swelled to many times its original volume. Another characteristic observed by comparing lower-temperature samples to the samples shown in Figure 3 is that the cracks appear only in regions where all of the matrix aluminum has been consumed in forming the $U(Al,Si)_3$ interaction product. When aluminum is present, it reacts with the U_3Si_2 fuel particles in an

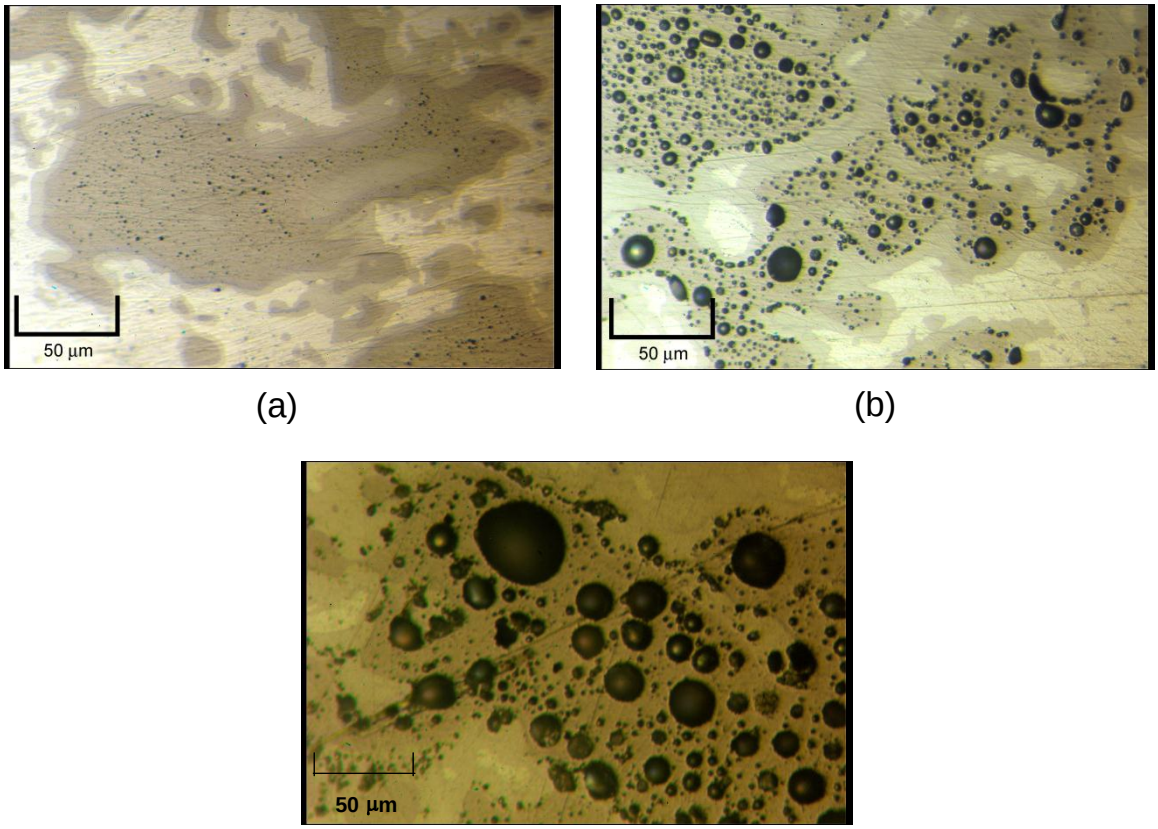


FIGURE 1 Optical Micrographs of Plate U0R040 Irradiated in the RERTR-8 Test in ATR: (a) Sample Where T = 105°C and FD = 3.2×10^{21} f/cm^3; (b) Sample Where T = 136°C and FD = 5.4×10^{21} f/cm^3; (c) Sample Where T = 160°C and FD = 6.1×10^{21} f/cm^3

irradiation-enhanced diffusion process to form $U(Al,Si)_3$ where the Al/Si ratio is about two. Once the aluminum has been depleted, the Al/Si ratio was found to be much less than two, and it is hypothesized that further diffusion leads to stresses and cracking as other phase compositions form. Fission gas then fills and expands these spaces, which would result in breakaway swelling in plate-type fuels. The dataset is sparse, and the uncertainty in the irradiation temperatures is estimated to be ±50°C, so only limited quantitative information can be deduced. However, it is clear from examination of other HANS-3 specimens that fracturing occurs only after the aluminum is consumed; the threshold temperature appears to be in the range of 200–300°C, or perhaps >250°C based on examination of two nominally 250°C samples, one of which exhibited the phenomenon and the other of which did not. Owing to the absolute importance of avoiding such failures, once this mechanism was identified in 1995, the Reduced Enrichment for Research and Test Reactor (RERTR) program has recommended conservatively that fuels expected to be used under such conditions be limited in burnup to that which results in the matrix aluminum volume fraction reaching 15 vol%. The implications for ^{99}Mo targets will be discussed in the next section.

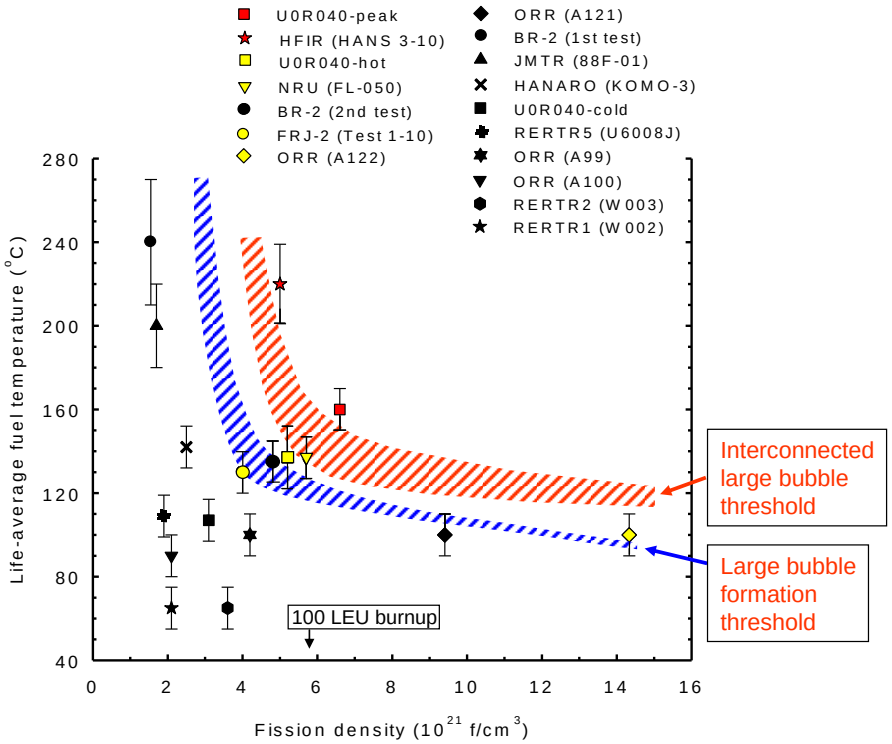
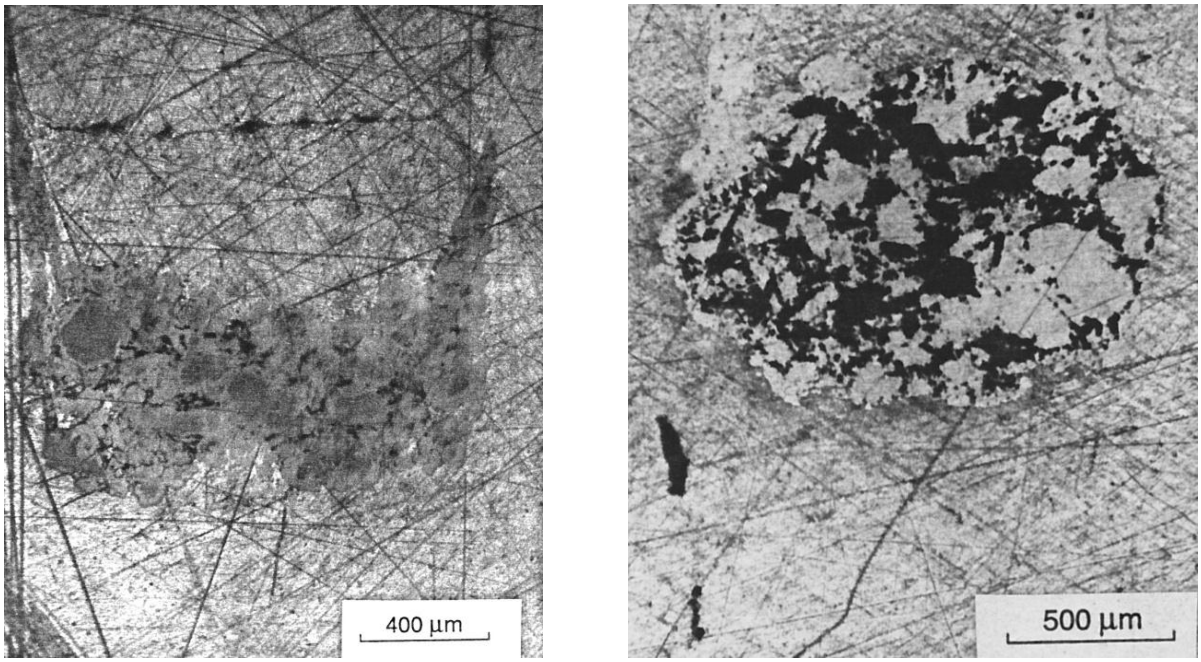


FIGURE 2 Fission Density-Temperature Threshold for Large Bubble Formation (greater than 5 µm) and Threshold for Interconnected Gas Bubble Formation in U_3Si_2 (The shaded areas indicate the uncertainty in the thresholds.)

In addition to qualification with respect to irradiation performance, a fuel must be qualified by its commercial fabricator with respect to its ability to fabricate the fuel to meet the specifications required by the customer. As indicated in the last section, one manufacturer has demonstrated its ability to manufacture U_3Si_2 dispersion fuel up to loading of 6.0 gU/cm^3. However, no manufacturer has qualified its fabrication process using U_3Si_2 powder with a content of fine (≤44 mm) particles of more than 50 wt%; the significance of this fact will become clear in the next section.



(a) (b)

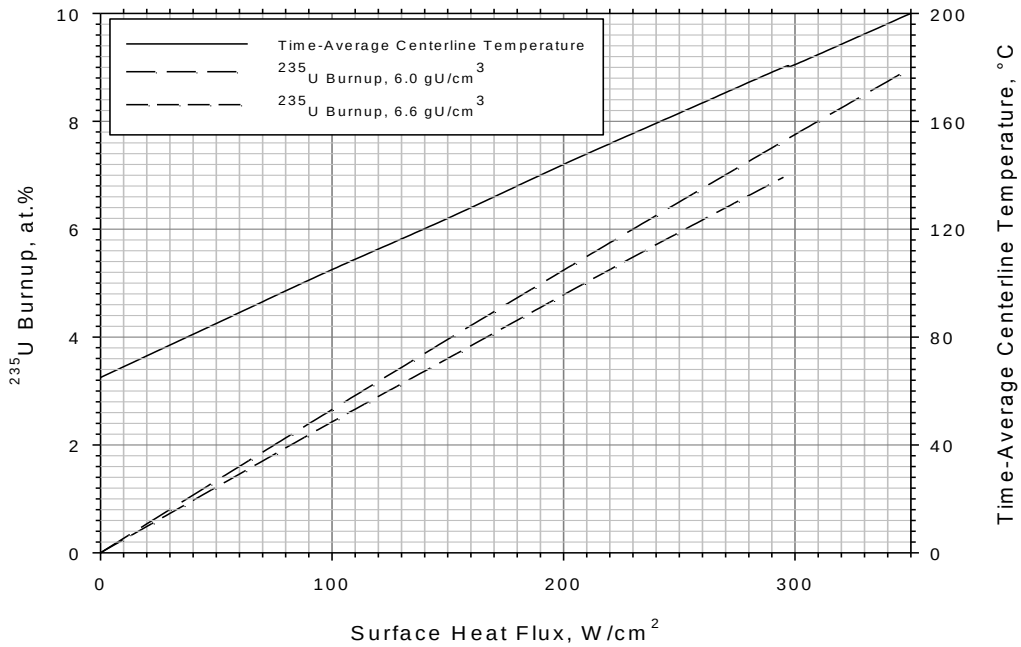
FIGURE 3 Optical Micrographs of Samples Irradiated in the HANS-3 test in HFIR: (a) Specimen 18, Where T = 250°C and FD = 4.0×10^{21} f/cm^3; (b) Specimen 17, Where T = 425°C and FD = 3.7×10^{21} f/cm^3

3 QUALIFICATION OF AND LIMITS FOR U_3Si_2 DISPERSION FUEL FOR ^{99}Mo TARGETS

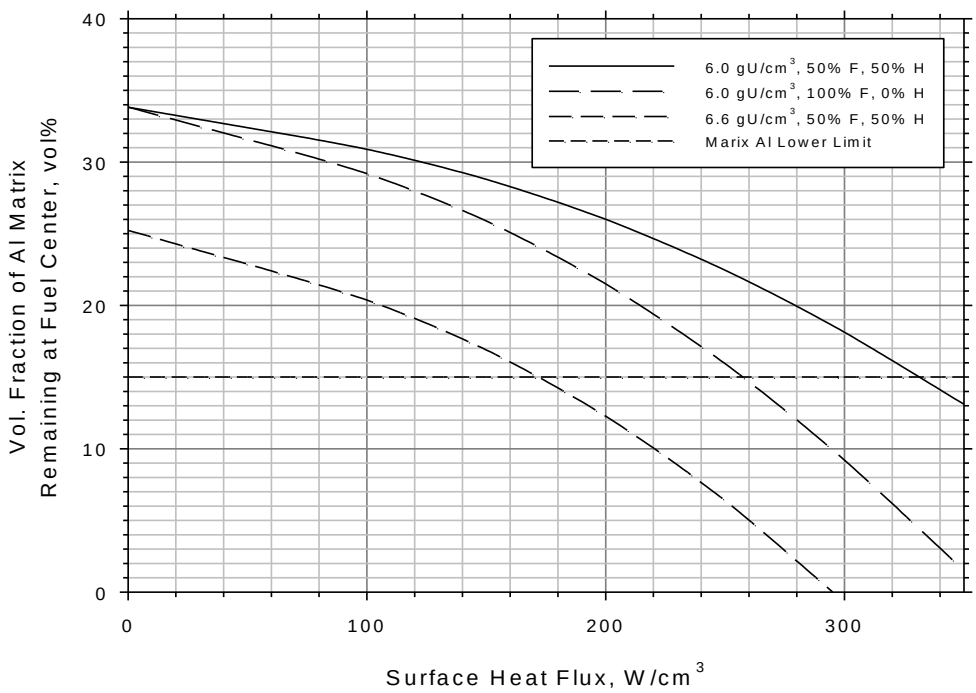
Figure 4 shows the results of the simulated irradiation of a U_3Si_2 dispersion target irradiated for 6.5 days in a test reactor with water chemistry like that of the HFR- (High Flux Reactor) Petten. The simulation was performed using a much-simplified version of the PLATE code (Hayes et al. 2002). An interesting aspect of this simulation study is that it explores the feasibility of using different mixtures of fine (diameter ≤44 μm) and heavy (≤125 μm) U_3Si_2 fuel particles for during target fabrication. The use of more fine particles than normally used to fabricate U_3Si_2 dispersion fuels could enhance the dissolution of the irradiated target by increasing the amount of interaction product produced. The irradiation parameters are typical for at least one of the LEU plate targets being considered by a commercial ^{99}Mo producer.

To evaluate the feasibility of using U_3Si_2 targets, one must consider the two failure phenomena discussed in the preceding section. With respect to the maximum bubble size in the fuel particle criterion, Figure 2 indicates that as long as the fission density remains below 2.2×10^{21} fissions/cm^3 s (and as long as the matrix aluminum has not been completely consumed), breakaway swelling will not occur, regardless of the irradiation temperature. Maximum fission density in a ^{99}Mo target is not expected to exceed 5×10^{20} fissions/cm^3 s (~10% burnup), so all U_3Si_2 dispersion targets are expected stay well below the limit set by the bubble-size criterion.

However, with respect to the criterion of avoiding total matrix aluminum consumption, Figure 4 shows that the amount of matrix aluminum drops below the recommended conservative limit of 15 vol% for the higher-heat flux cases. The simulations also showed that even when all of the matrix aluminum was consumed in the cases where 100 wt% fine fuel particles are used, a significant amount of unreacted fuel remains. Moreover, it is impossible to fabricate a dispersion fuel plate in which the fuel is homogeneously distributed throughout the fuel meat. Therefore, one must look at the uranium distribution allowed by the fuel specification, which for target-sized plates might be set at ±10% in a 1 cm^2 area. As one can see, raising the fuel density to 6.6 gU/cm^3 would significantly reduce the allowable surface heat flux. It must be emphasized that because of the underlying uncertainty in irradiation behavior data under high-temperature conditions and because of the simplicity of the calculational model used in the simulation, the results of the simulation must be applied very conservatively. Certainly, additional studies using the latest version of the PLATE code should be done to better define the expected behavior of targets for use under the anticipated irradiation conditions. On the other hand, the low burnup of ^{99}Mo targets could be a significant mitigating factor; the few data points available under very-high-temperature conditions came from samples having very-high burnup, where the quantity of fission gas produced could be almost an order of magnitude higher than in a target.



(a)



(b)

FIGURE 4 Results of Simulated Irradiation of 6.0-gU/cm^3 LEU U_3Si_2 Target at Various Power Densities: (a) Time-Average Centerline Temperature and ^{235}U Burnup; (b) Volume Fraction of Matrix Aluminum Remaining (The plots for 6.6.gU/cm^3 show the effect of a 10% fuel overload at the center of the target.)

4 CONCLUSION

While this evaluation has shown that sufficient data are available to consider low-enriched, 6.0 gU/cm^3 U_3Si_2 dispersions qualified for use as ^{99}Mo targets when surface heat fluxes are no higher than 150-200 W/cm^2, additional work would be necessary to justify their use under higher-heat flux conditions. First, more sophisticated calculations using the latest version of the PLATE code should be made to obtain better estimates of expected irradiation behavior where these high heat fluxes are needed. Since some mitigating factors are expected in the PLATE calculations, it might be possible to raise the heat-flux limit for which the current fuel could be considered to be qualified. No matter what this heat flux value is, irradiation and destructive postirradiation examination of lead test targets would be required to validate these conclusions.

For even higher-heat flux conditions, a significant irradiation program would be required to provide sufficient data for qualification. Should a fines content greater than 50 wt% be desired, manufacturing qualification will also be required.

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